Kinetics of Direct Oxidation of H_2S in Coal Gas to Elemental Sulfur

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OBJECTIVES

Direct oxidation of H_2S to elemental sulfur in the presence of SO_2 is ideally suited for coal gas from commercial gasifiers with a quench system to remove essentially all the trace contaminants except H_2S. This direct oxidation process has the potential to produce a super clean coal gas more economically than both conventional amine-based processes and the hot-gas desulfurization using regenerable metal oxide sorbents followed by Direct Sulfur Recovery Process. The main objective of this research is to support the near- and long-term process development efforts to commercialize this direct oxidation technology. The objectives of this research are to measure kinetics of direct oxidation of H_2S to elemental sulfur in the presence of a simulated coal gas mixture containing SO_2, H_2, and moisture, using 160-μm C-500-04 alumina catalyst particles and a micro bubble reactor, and to develop kinetic rate equations and model the direct oxidation process to assist in the design of large-scale plants. This heterogeneous catalytic reaction has gaseous reactants such as H_2S and SO_2. However, this heterogeneous catalytic reaction has heterogeneous products such as liquid elemental sulfur and steam.

Experiments on conversion of hydrogen sulfide into liquid elemental sulfur were carried out for the space time range of 0.059 – 0.87 seconds at 125 - 155^{o}C to evaluate effects of reaction temperature, H_2S concentration, reaction pressure, and catalyst loading on conversion of hydrogen sulfide into liquid elemental sulfur. Simulated coal gas mixtures consist of 62 - 78 v% hydrogen, 3,000 – 7,000-ppmv hydrogen sulfide, 1,500 - 3,500 ppmv sulfur dioxide, and 10 vol % moisture, and nitrogen as remainder. The volumetric feed rate of a simulated coal gas mixture to a micro bubble reactor is 50 cm^3/min at room temperature and atmospheric pressure. The temperature of the reactor is controlled in an oven at 125 - 155^{o}C. The pressure of the reactor is maintained at 40 - 170 psia.

ACCOMPLISHMENTS TO DATE

- Reaction temperature affects conversion of H_2S to elemental sulfur, but conversion of H_2S to elemental sulfur does not follow the Arrhenius' equation.

- Reaction pressure affects significantly conversion of H_2S to elemental sulfur in the pressure range of 40 -170 psia.

- A reaction rate equation for the conversion of H_2S to element sulfur in the presence of SO_2 over the total reaction pressure range of 40 – 120 psia was developed with the surface reaction mechanisms as follows. Gaseous hydrogen sulfide is predominantly attached to active sites on the surface of catalyst particles, and then the attached H_2S is reacted with gaseous SO_2 from a bulk gaseous reaction mixture to produce liquid elemental sulfur and water. Water on active sites, produced from the reaction of H_2S with SO_2, is mostly evaporated into the gaseous bulk reaction mixture.

- The developed reaction model suggests that H_2S is strongly adsorbed onto active sites of catalyst particles in the preference over SO_2, and the reaction for conversion of H_2S to elemental sulfur is second order with respect to partial pressure of H_2S and first order with respect to partial pressure of SO_2 over the total reaction pressure range of 40 – 120 psia .

FUTURE WORK

Reaction kinetics on conversion of both hydrogen sulfide to elemental sulfur and carbon monoxide to carbonyl sulfide will be investigated in the presence of a reduced gas mixture of hydrogen and carbon monoxide with pellet catalyst paricles in comparison with honeycomb catalysts.

PUBLICATIONS AND PRESENTATIONS

Kinetics of Direct Oxidation of Hydrogen Sulfide in Coal Gas to Elemental Sulfur, Kyung C. Kwon, Santosh K. Gangwal, Janelle C. Houston, and Erica D. Jackson, DOE Annual Contractors' Review Meeting, Pittsburgh Marriott City Center, Pittsburgh, PA, June 4 – 5, 2002

Conversion of Hydrogen Sulfide in Coal Gas to Elemental Sulfur, Kyung C. Kwon, Santosh K. Gangwal, Suresh C. Jain, YoonKook Park, Janelle C. Houston and Erica D. Jackson, AIChE Annual Meeting, Indiana Convention Center, Indianapolis, Indiana, November 3 – 8, 2002.

Kinetics of Direct Oxidation of Hydrogen Sulfide in Coal Gas to Elemental Sulfur, Kyung C. Kwon, Santosh K. Gangwal, and Erica D. Jackson, DOE Annual Contractors' Review Meeting, Pittsburgh Marriott City Center, Pittsburgh, PA, June 3 – 4, 2003

Conversion of H_2S in Coal Gases to Liquid Elemental Sulfur in a Micro Bubble Reactor, Kwon, K. C., YoonKook Park, S. K. Gangwal, Suresh Jain, and Erica Jackson, Engineered Particle Systems: Synthesis, Processes and Application Topical Proceedings, AIChE Annual Meeting, San Francisco, CA, November 16 -21, 2003.

Conversion of H_2S in Coal Gas to Liquid Elemental Sulfur in a Micro Bubble Reactor, Kyung C. Kwon, Santosh K. Gangwal, Suresh C. Jain, YoonKook Park, and Erica D. Jackson, Presented at AIChE Annual Meeting, San Francisco, CA, November 16 -21, 2003.

Oxidation of H_2S in Coal Gas to Liquid Elemental Sulfur in a Micro Bubble Reactor, Kyung C. Kwon, Suresh C. Jain, YoonKook Park, Monica I McCoy and Iisha Griffin, Presented at AIChE Annual Meeting, Austin, TX, November 7 – 12, 2004.

AWARDS RECEIVED AS A RESULT OF SUPPORTED RESEARCH

Conversion of Hydrogen Sulfide in Coal Gases to Elemental Sulfur with Monolithic catalysts: DE-FG26-04NT42129.

STUDENTS SUPPORTED UNDER THIS GRANT

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